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## PROVISIONAL APPLICATION COVER SHEET

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APPARATUS FOR TREATING SOLUTIONS OF HIGH OSMOTIC STRENGTH						
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## APPARATUS FOR TREATING SOLUTIONS OF HIGH OSMOTIC STRENGTH

Field of the Invention

The present invention pertains to an apparatus and method for treating a solution of high osmotic strength, especially seawater, by passing the solution through a vessel containing spiral wound reverse osmosis or nanofiltration elements. In particular, the apparatus and method allow for a more even distribution of flux within the vessel. Advantageous performance properties compared to conventional methods may be obtained, including higher vessel productivity, increased recovery, lower fouling rate, and lower required applied pressure.

Background of the Invention

Osmosis is the process whereby solvent passes through a semi-permeable membrane and moves from a solution of low solute concentration to one of high solute concentration, diluting the latter. In reverse osmosis (RO), pressure is applied to the high solute concentration side of the membrane and the chemical potential gradient that drives osmosis is reversed. The result is flow of solvent across the membrane, from high solute concentration to lower solute concentration, which allows a purified solvent solution to be produced. Reverse osmosis is now commonly used to create potable water from seawater.

Desalination of water by reverse osmosis has been practiced commercially since the introduction of cellulose acetate membranes by Loeb and Sourirajan. However, in the early 1980's, FilmTec Corporation began producing aromatic polyamide membranes with substantially improved flux and salt passage. These membranes were created in an interfacial polymerization to form an aromatic polyamide, as disclosed by Cadotte (US 4,277,344). Today, virtually all commercial membranes for seawater desalination are based on this process, and reverse osmosis accounts for about 40% of global desalination.

Nanofiltration (NF) is similar to reverse osmosis in that pressure applied to the membrane overcomes an osmotic pressure difference and forces water through a membrane. However, nanofiltration membranes are distinguished by the fact that some salts are substantially passed, while other salt are selectively retained. NF is most commonly applied to feed streams having low salt concentrations, but it has also been used to selectively remove components from seawater. For instance, US 6508936 describes use of NF as a pretreatment that may advantageously remove hardness ions from seawater.

US 4723603 describes use of NF as a means to reduce sulfate ions in seawater prior to downhole injection. While NF membranes do not suffice for total desalination, the low net osmotic pressure difference resulting from their high passage of NaCl is advantageous for specific applications.

5 RO and NF membranes are typically configured in a series of spiral wound elements because such elements allow a large amount of membrane area to be packed into a small volume. A classic spiral wound element design is shown in Figure 1. "Feed" liquid flows axially through a feed spacer sheet and exits on the opposite end as "concentrate". "Permeate" passes under pressure through membrane envelopes and is directed to a  
10 permeate collection tube by a permeate carrier sheet. Spiral wound elements and corresponding vessels are commercially available in a variety of standard diameters (e.g. 4.5, 6.3, 10, 15, 20 cm), but a one meter long element with a nominal 20 cm diameter is currently the standard for large systems. For seawater applications, each 20 cm diameter element typically contains between 26.5 m<sup>2</sup> (285 ft<sup>2</sup>) and 35.3 m<sup>2</sup> (380 ft<sup>2</sup>) of active  
15 membrane area.

Spiral wound elements are generally placed inside of a cylindrical pressure vessel for operation. In commercial RO applications, a large filtration system may be composed of more than 10,000 elements, usually distributed in pressure vessels containing  
20 4 to 8 elements each. The pressure vessels have ports for inputting the pressurized feed solution and removing the concentrate and permeate solutions. Feed flows axially through each of the elements in series. By connecting the permeate collection tubes of different elements, the effect is to create one long element in a vessel.

Pressure vessels can be further combined in series or parallel with other pressure vessels to create a membrane filtration system. Commonly, vessels are cascaded in a  
25 tapered arrangement, so that the combined concentrate from several parallel vessels in a stage becomes the feed to a successive stage composed of a smaller number of parallel vessels. Within a stage, the number of parallel vessels, the number elements within a vessel, and the type of element can all be selected to optimize performance. System design can be further optimized by incorporating a variety of other options, including recirculation  
30 loops, booster pumps between stages, and permeate pressurization. Additionally, permeate may be removed from either or both ends of a vessel and restrictions to permeate flow (between elements or within a permeate tube) may be used to balance fluxes and selectively

remove permeate. These and other options allow a designer to optimize system performance while adhering to practical constraints on element operation.

Guidelines established by element manufacturers are among the primary constraints considered in system design. Examples of such constraints can be found in literature published by manufactures of RO elements, such as ("Membranes System Design Guidelines for 8-inch FILMTEC™ Elements", Form No. 609-21010-702XQRP, FilmTec Corp., Edina, MN, 12/18/03). Typically, an upper bound on applied pressure, a maximum feed flow rate, a minimum concentrate flow rate, and a maximum recovery (volume of permeate divided by volume of feed) for an element may all be specified. For example, operating limits for a FILMTEC™ SW30HR-380 seawater element using open intake include <69 bar (<1000 psi) applied pressure, <338 m<sup>3</sup>/d (<62 gpm) feed flow, and >98 m<sup>3</sup>/d (>18 gpm) concentrate flow rate. Other manufacturers have established similar guidelines, although a maximum element recovery often replaces feed and concentrate flow specifications, and there has recently been a move to allow higher applied pressures in seawater systems.

Constraints on flux are also common and particularly relevant to the present invention. Flux is defined as the permeate water volumetric flow rate per unit of area. Highflux increases concentration polarization and polarization decreases permeate quality. Further, it is well known that high flux can increase fouling, resulting in the need for more frequent cleaning. This relationship between flux and fouling has been studied extensively, and flux guidelines are commonly established based on the fouling potential of different waters. Among the most common methods of characterizing fouling potential is the Silt Density Index (SDI), and this value is used to establish limits on maximum flux for seawater applications.

Membrane manufactures commonly publish design guidelines for maximum flux. FilmTec specifies a maximum average flux for seawater elements operated on low SDI water (commonly well or MF pre-treated water) of 39 L/m<sup>2</sup>/hr (23 gfd). For an open intake seawater system (SDI < 5), FilmTec's specified design guidelines correspond to 34 L/m<sup>2</sup>/hr (20 gfd). In design guidelines from TORAY, the maximum flow specified for seawater elements (based on nominal areas) corresponds to 35.3 L/m<sup>2</sup>/hr (20.8 gfd) for beach wells and 28.9 L/m<sup>2</sup>/hr (17.0 gfd) for open inlet ("Design Guidelines", TORAY Membranes America, Inc., San Diego, CA).

There are other means to create design guidelines that limit flux through the membrane elements. For instance, Hydranautics' design limits specify an average system flux of 13-24 L/m<sup>2</sup>/hr (8-14 GFD) for surface water (SDI 2-4). More specifically to seawater, one of their recent publications (*Desalination*, 135, 2001, 61-68) states,

5 "The average permeate flux in seawater systems is maintained at relatively low values: 7-8 gfd for surface water feed and 10 gfd (16.8 l/m<sup>2</sup>-hr) for seawater from beach wells. The difference in flux rates between the two water source types results from better quality of the well water and therefore, a lower fouling rate of the membranes. These flux values are relatively low and only about 50% of the permeate flux values used in brackish RO

10 systems. Attempts to operate seawater systems at higher flux rates have usually resulted in irreversible flux decline." That paper extols the economic benefit of improved pretreatment, producing lower SDI values and allowing operation of a 55% recovery system at an average flux of 19 L/m<sup>2</sup>/hr (11 gfd).

Figure 2 shows that specifying a limiting average system flux of around

15 12-17 L/m<sup>2</sup>/hr (7-10 gfd) is not inconsistent with specifying a maximum element flux. This graph, described in more detail in Comparative Example III, is based on a typical seawater configuration using 6 elements per vessel and 50% volume recovery of the feed. The graph indicates that an average flux of 31 L/m<sup>2</sup>/hr (18 gfd) for the lead element corresponds to a flux at the entrance to that element of 34 L/m<sup>2</sup>/hr (20 gfd) and an average

20 system flux of 17 L/m<sup>2</sup>/hr (10 gfd). While there is not absolute agreement between the different methods for specifying limits on flux, and the relative agreement between methods depends on operating conditions (e.g. salinity, temperature, recovery, pressure), the need for such a flux limitation in system design is universally recognized. It is currently a common practice in bids to design seawater systems with both a 30 to 34 L/m<sup>2</sup>/hr (18 to 20 gfd)

25 maximum lead element flux and an average system flux, commonly around 13 to 15 L/m<sup>2</sup>/hr (8 to 9 gfd).

Figure 2 also illustrates a problem for attaining higher recoveries in seawater RO operations. The flux decreases from the inlet end of the vessel (34 L/m<sup>2</sup>/hr) to the outlet end (5 L/m<sup>2</sup>/hr) as the rejected water concentrates and the feed osmotic pressure increases.

30 In reverse osmosis, the flux through a membrane is essentially proportional to a net driving pressure. Net driving pressure is calculated by subtracting both the permeate pressure and the osmotic pressure difference across the membrane from the applied pressure. In the absence of polarization, the osmotic pressure difference across highly rejecting membranes is approximately equal to the osmotic pressure of the feed solution. For a typical salinity of

3.5% the osmotic pressure at the inlet port (before the lead element) would be about 26 bar (about 380 psi). At 40% recovery, the concentrate would then have about 5.8% salinity and the osmotic pressure at the outlet (after the last element) would be about 44 bar (about 630 psi). The increase in osmotic pressure for the feeds to successive elements dramatically decreases net driving pressure and flux for downstream elements. This problem is further aggravated by hydraulic resistance to feed flow within each element, resulting in a pressure drop down the vessel. Both aspects, but particularly the former in the case of high osmotic strengths solutions like seawater, combine to result in flux imbalance.

Flux imbalance contributes to fouling and decreases overall water quality due to polarization in the upstream elements and low flux in the downstream elements. Historically, these problems have been mitigated by the use of elements with low specific flux for high osmotic strength applications. The specific flux of a membrane is defined as the flux (permeate volumetric flow rate per unit area of membrane) divided by the net driving pressure, as calculated above. In this way, high pressure can be applied to the first element in series while flux from that element is kept within guidelines. The high applied pressure allows the change in osmotic pressure from the lead end of the vessel to the tail end to be maintained at a relatively small fraction of the initial net driving pressure. Table 1 shows standard specific flux values estimated for four FilmTec seawater elements from the manufacturer's specified flow, area, and test conditions.

Table 1. Standard specific flux of FilmTec seawater elements

FILMTEC™ Element	Flow m <sup>3</sup> /d (gpd)	Reject (%)	Area m <sup>2</sup> (ft <sup>2</sup> )	Standard Specific Flux l/m <sup>2</sup> /hr/bar (gfd/psi)
SW30HR-380	22.7 (6000)	99.7	35.3 (380)	0.097 (0.039)
SW30HR-320	22.7 (6000)	99.75	29.7 (320)	1.15 (0.047)
SW30HR LE-380	28.4 (7500)	99.75	35.3 (380)	1.22 (0.049)
SW30-380	34.0 (9000)	99.4	35.3 (380)	1.46 (0.059)

Table 2 lists manufacturers' literature values for flow and salt passage for several one meter long, 20 cm diameter commercial seawater elements. It is noteworthy that the flows associated with commercial seawater offerings are fairly consistent between manufacturers. Using an active membrane area of 32.5 m<sup>2</sup> (350 ft<sup>2</sup>), the standard specific flux for elements in Table 2 would all fall between 0.74 L/m<sup>2</sup>/hr/bar (0.03 gfd/psi) and

1.23 L/m<sup>2</sup>/hr/bar (0.05 gfd/psi). A Hydranautics publication shows that 1.0 L/m<sup>2</sup>/hr/bar (0.04 gfd/psi) for seawater elements has been the norm since commercial introduction of aromatic polyamide membranes in the mid 80's (M. Wilf & K. Klinko, "Improving performance and economics of RO seawater desalting using capillary membrane pretreatment", Hydranautics, Inc., June 1998). Because elements for seawater desalination are commonly operated with between about 55 bar (800 psi) and 69 bar (1000 psi) applied pressure, with an osmotic strength of the feed usually between 24 bar (350 psi) and 31 bar (450 psi), this 1.0 L/m<sup>2</sup>/hr/bar (0.04 gfd/psi) value prevents substantially over-fluxing the lead element. By contrast, substantially higher specific fluxes have been commercially available for non-seawater applications since before 1990. In fact, over the last decade the specific flux for commercial brackish water elements has more than doubled.

Table 2. Flux and rejection for commercial seawater elements

Manufacturer	Element	Flow m <sup>3</sup> /d (gpd)	Reject (%)
TORAY Industries, Inc	TM820-370	23.0 (6076)	99.75
TORAY Industries, Inc	TM820-400	25.0 (6604)	99.75
TORAY Industries, Inc	SU-820	16.0 (4227)	99.8
TORAY Industries, Inc	SU-820L	20.0 (5283)	99.7
TORAY Industries, Inc	SU-820FA	19.0 (5019)	99.75
TORAY Industries, Inc	TM820H-370	21.0 (5548)	99.8
Hydranautics	SWC1	18.9 (5000)	99.6
Hydranautics	SWC3	22.3 (5900)	99.7
Hydranautics	SWC3+	26.5 (7000)	99.8
Hydranautics	SWC4	20.8 (5500)	99.8
Hydranautics	SWC4+	24.6 (6500)	99.8
Koch Membrane Systems, Inc.	TFC®2822 SS-300	18.9 (5000)	99.6
Koch Membrane Systems, Inc.	TFC®2822 SS-360	22.7 (6000)	99.6
Koch Membrane Systems, Inc	TFC®2822 SS-300 Premium	17.0 (4500)	99.75
Koch Membrane Systems, Inc.	TFC®2822 SS-360 Premium	20.8 (5500)	99.75
TriSep Corporation	8040-ACMS-SSA	22.7 (6000)	99.5

In addition to limiting the standard specific flux of seawater elements, another means to address the problems of flux imbalance has been to limit the recovery of seawater systems. Conventional seawater systems are commonly designed to operate with a recovery of around 40-45%, so that flux limitations are not exceeded. However, it is known that total system cost is integrally tied to recovery. Higher recovery means that less water must undergo pretreatment, and this has impact on both operating and capital expense. Similarly, higher recoveries can reduce costs for the RO portion of a system, particularly in components that increase in number with plant size (e.g. piping, electrical, elements and vessels). Also, increased recovery means less water must be raised to high pressure, and this minimizes economic loss from natural inefficiencies in energy recovery during operation. Finally, higher recoveries may reduce the volume of plant discharge. However, the recoveries which can be obtained in a vessel, and the associated benefits, have

conventionally been limited by the threats of fouling at the lead end and low flow (for both permeate and concentrate) at the tail end.

Increasing the number of elements within a vessel provides one means to maximize the recovery for that vessel while preventing overfluxing of the first element. Of course, this approach is only generally applicable for new installations where the length of vessels is not set, and it is not easily used to increase recoveries at existing facilities. Also, even in the case of new installations, this solution generally results in a situation where the last elements in a vessel operate at very low flux. Because less water passes through these downstream elements, but the rate of salt diffusion is similar to that for other elements, their permeate quality is lower. Additionally, the cost for such systems is increased due to a larger number of elements and the need for longer vessels.

In the last few years, several other examples have been cited for improving seawater economics while adhering to flux guidelines. US 6187200 describes systems comprising multiple stages with backpressure on the first stage or an inter-stage boost between stages. While these methods have some advantages, they require additional cost in pumps, plumbing, and extra pressure vessels. Further, these two-stage systems are often designed to operate with at least one stage at very high pressures, and this makes equipment more expensive and has an impact on element components (US 6277282). Alternatively, improved pre-treatment of seawater has been advocated to allow for higher average flux guidelines and greater tolerance for flux imbalance. This also requires additional capital expense (this time for pretreatment), and the impact pretreatment will have on fouling is difficult to quantify a priori. Finally, very efficient energy recovery devices have been used to make substantially lower recoveries more economically competitive. As mentioned previously, lower recoveries would limit flux imbalance within a seawater system. However, this method involves additional equipment for energy recovery, potentially increased costs due to a greater volume of pretreated water, and reliance on new technologies that may not be fully embraced in the market.

Over the last two decades, reverse osmosis has grown to account for about 40% of the global production of desalinated seawater. However, the rate of additional growth has potential to be dramatically impacted by improved economics. Chief among the current shortcomings in the present state of the art is susceptibility to fouling, a problem made more extreme by operation at high fluxes. Improvements are needed which allow for operation at higher average flux, operation at higher recoveries within a vessel, and operation with a

more uniform flux distribution. System design with increased vessel efficiency may substantially reduce capital expenses. Additionally, it would substantially benefit operating costs for systems to be able to provide similar flows with lower applied pressure.

The disclosed invention addresses these issues and also provides a means by which existing reverse osmosis systems may be inexpensively upgraded.

#### Summary of the Invention

Purification of seawater can be performed by conventional methods using a vessel of reverse osmosis elements in series. However, the decrease in flux that results from concentrating the feed in successive elements substantially limits the recoveries that may be obtained. Additionally, there are several problems with this inhomogeneous utilization of membrane. For upstream elements, high flux can substantially shorten the life of an element due to fouling and scaling. High flux also promotes concentration polarization and polarization decreases the effective rejection of the membrane. Lower flux in downstream elements is also undesirable, not only because of decreased productivity, but also because lower flux means higher solute concentration in the permeate.

It is a primary objective of the present invention to provide a spiral wound membrane system that allows seawater to be purified within a single vessel under conditions that provide advantageous performance properties for the vessel while not exceeding flux guidelines. The advantageous performance properties may manifest themselves in lower capital costs, higher recovery, greater vessel productivity, lower fouling rates, and/or lower required applied pressure, depending on the method of operation.

A fundamental aspect of this invention is the combination within a vessel of at least three connected elements in series, where the standard specific fluxes differ substantially between elements to provide a more even flux distribution. In this invention, the standard specific fluxes within the vessel differ by at least a factor of 1.5, and more preferably by at least a factor of 2. In one embodiment, a feed solution with osmotic pressure of at least 20 bar is treated using a filtration vessel that contains a downstream element having a standard specific flux that is least 1.5 times that of the lead element. In another embodiment, the standard specific flux for the tail element divided by the standard specific flux for the lead element is great that 2, and a third element has a standard specific flux that is between 1.25 and 1.75 times that of the minimum standard specific flux. In still another embodiment, a barrier to permeate flow within the filtration vessel defines two separate permeate streams that leave the vessel at opposite ends. Most preferably, the vessel's lead

element has a high standard specific flux, but the combined permeate stream from upstream elements is caused to become the feed to a second pass, so as to create a more even flux distribution within the first pass vessel.

This invention is expected to have greatest utility in seawater desalination.

- 5 Combining elements having widely differing standard specific fluxes within a vessel allows flux guidelines to be maintained while operating at higher recoveries or lower energies, as compared to conventional systems. This is best achieved for seawater when a downstream element (preferably the tail element) has a standard specific flux that is at least 1.5 L/m<sup>2</sup>/hr/bar. It is also often desirable that an upstream element (preferably the lead  
10 element) has a standard specific flux less than 1.0 L/m<sup>2</sup>/hr/bar.

- Another embodiment of our invention entails using different feed spacer cross sectional areas in upstream and downstream elements. This aspect supports the higher recoveries enabled with our invention. Despite decreases in feed volume down the vessel, a 15% or 30% decrease in feed spacer cross sectional area allows velocities in the feed  
15 channel to be maintained when elements are operated according to a 15% recovery guideline.

- Our invention also includes selectively incorporating different types of feed spacer sheets within specific elements in a vessel. Due to the high salt concentrations at recoveries enabled by our invention, it is preferred that downstream elements use a feed spacer that  
20 produces increased turbulence but generates greater pressure drop, as compared to that in the first element in series. Most preferably, the tail element has a standard pressure gradient greater than 0.4 bar/m and/or its standard pressure gradient is at least 50% greater than the standard pressure gradient for the first element in series.

- As mentioned, this invention preferably makes use of a downstream element having  
25 a standard specific flux of at least 1.5 L/m<sup>2</sup>/hr/bar. At the same time, it is desired that this element maintain a low standard solute permeability. One embodiment requires that the last element in series produce a permeate salt concentration of less than 500 ppm when tested using 25°C, 32000 ppm NaCl in the feed, 8% recovery, and a flux of 27 L/m<sup>2</sup>/hr. In another embodiment, both the upstream and downstream elements have an NaCl passage at these  
30 conditions of greater than 20%, but a sulfate passage of less than 1% is obtained for both elements after the 2000 ppm MgSO<sub>4</sub> is added to the 32000 ppm NaCl.

It is another primary aspect of the present invention to use an apparatus with the aforementioned qualities to purify water, wherein the difference in applied pressure and

osmotic pressure at the vessel's inlet is at least twice that at the vessel's outlet.

Our arrangement of elements differing in standard specific flux allows a more even distribution of flux between elements in the vessel, and one embodiment provides for an average vessel flux that is not less than half the first element flux. A related embodiment  
5 uses a high average vessel flux ( $>20 \text{ L/m}^2/\text{hr}$ ) while the first element flux is maintained within typical design guidelines ( $<34 \text{ L/m}^2/\text{hr}$ ), enabling higher system recoveries.

In another embodiment, an average vessel flux is at least 70%, more preferably 80%, of the lead element flux, while the lead element is kept to less than  $25 \text{ L/m}^2/\text{hr}$ , so as to further avoid fouling. Our invention is particularly advantageous when the osmotic pressure of the  
10 feed solution exceeds 20 bar at the vessel inlet, so that concentration of the reject solution would normally dramatically reduce flow of the last element.

The invention also includes a process whereby an existing vessel containing only elements with standard specific flux less than  $1.25 \text{ L/m}^2/\text{hr}/\text{bar}$  is upgraded, allowing either greater recovery or operation at lower pressure. In this case, at least one initial element is  
15 removed and another with at least  $1.5 \text{ L/m}^2/\text{hr}/\text{bar}$  is added to the vessel.

#### Brief description of the drawings

This invention and its preferred embodiments may be better understood through reference to the detailed description of the invention, accompanied by figures described below. Within these sections, like reference numerals refer to like elements.

20 FIG. 1 is a perspective, partially cutaway view of a spiral wound element. The element is formed by alternately wrapping filtration envelopes and feed spacer sheets about a central permeate collection tube. The filtration envelop comprises a permeate carrier sheet sandwiched between two sheets of membrane.

FIG. 2 is a graph showing how permeate flux may substantially decrease from the  
25 lead end to the tail end of the vessel. The figure presents calculated flux for a six element vessel corresponding to operating conditions detailed in Comparative Example III.

FIG. 3 is a schematic that shows a typical configuration for a vessel containing at least three elements in series.

FIG. 4 is a schematic that shows another typical configuration for a vessel  
30 containing at least three elements in series. In this figure, a barrier to permeate flow exists within the permeate tube of one element and permeate solution is removed from both ends of the vessel.

Detailed description of the invention

This invention focuses on improved use of spiral wound elements for purification of solutions having high osmotic strength. The construction of spiral wound elements has been described in more detail elsewhere (see US Patent Nos. 5,538,642 and 5,681,467 incorporated herein by reference). Referring to Fig. 1, the following brief description presents a typical spiral wound element. The element is formed by wrapping one or more membrane envelopes (2) and feed spacer sheet (4) about a central permeate collection tube (6). The envelopes (2) comprise two generally rectangular membrane sheets (8) surrounding a permeate carrier sheet (10). This "sandwich" structure is held together along three edges (14,16,18), while the fourth edge (20) of the envelope (2) abuts the permeate collection tube (6) so that the permeate carrier sheet (10) is in fluid contact with openings (22) in the permeate collection tube (6). Each envelope (2) is separated by feed spacer sheet (4) that is also wound about the collection tube (6). The feed spacer (4) is in fluid contact with both ends of the element (24,26) and it acts as a conduit for feed solution across the front surface (28) of the membrane (8).

In this invention, at least three spiral wound elements are encased within a cylindrical pressure vessel. Such pressure vessels are known in the art and are exemplified by US 6074595 and US 6165303. Referring to Fig. 3, a vessel (40) has ports (42,44) on opposite ends (46,48) for passing feed solution into the vessel and removing the concentrate solution. Within a vessel (40), elements are arranged in series. Feed solution flows from the lead element (50) at the inlet end (46) of the vessel (40), across intermediate elements (52), to the tail element (54) at the opposite outlet end (48) of the vessel (40). Brine seals (56) between elements and the vessel may be used to prevent this feed flow from bypassing elements. Interconnectors (58) used to connect permeate tubes (6) of adjacent elements, and the combined permeate is removed from at least one permeate port (62) in the vessel (40).

This invention is applicable to both individual vessels and parallel vessels within a membrane filtration system. In the art, it is understood that parallel vessels are vessels being supplied with essentially the same feed solution, being operated at essentially the same conditions, and containing the same number and type of elements. In this invention, elements of different types are used within a vessel, and two parallel vessels in that case must further contain the same type of element at each corresponding position within the parallel vessels. It is also understood in the art that elements of the same type are those supplied by the manufacturer with a separate designation corresponding to the group of

elements of that type. Elements of a given type have essentially the same construction (including feed spacers, membrane type, and area) and have a narrow range of acceptable flows and salt passage. For the elements in Tables 1 and 2, it is typical for the maximum deviation from the given flow to be specified as less than  $\pm 15\%$ .

5 This invention uses elements of differing standard specific flux. In defining the standard specific flux of an element, it can be noted that a specific flux for a membrane is commonly understood as the flux (permeate volumetric flow rate per unit area of active membrane) divided by the net driving pressure. While specific flux is often considered a constant of the membrane, it actually varies predictably as a function of temperature, concentration, and applied pressure. Additionally, the specific flux for any element  
10 operating in the field is subject to changes due to aging, fouling, and compaction. Hence, assigning a standard value to an element requires also specifying a specific point in time within the useful life of an element. For the purpose of this specification, the standard specific flux for an element is defined in terms of a test performed after 24 hours of initial operation, to allow for membrane equilibration, although subsequent changes to results of this test should be relatively small in the absence of substantial fouling. The standard  
15 specific flux for an element is determined using a test with 32000 ppm NaCl in the feed, 8% recovery, 25°C, and an average flux of 27 L/m<sup>2</sup>/hr (16 gfd). More specifically, the standard specific flux for an element is defined as the average flux (27 L/m<sup>2</sup>/hr) divided by a pressure term  $P$ , where  $P$  is calculated from quantities measurable during the test:  
20

$$P = (P_{\text{feed}} + P_{\text{conc}})/2 - P_{\text{perm}} - (\pi_{\text{FeedAvg}} - \pi_{\text{perm}})$$

$P_{\text{feed}}$  is the applied pressure on the inlet side of the element.  $P_{\text{conc}}$  is the applied pressure on opposite end (concentrate side) of the element, and  $P_{\text{conc}}$  is typically smaller than  $P_{\text{feed}}$  due to resistance to flow within the feed spacer (4).  $P_{\text{permeate}}$  is the applied pressure at the point  
25 where permeate exits an element, and this backpressure during the test is typically very small.  $\pi_{\text{FeedAvg}}$  is the osmotic pressure of a solution formed by mixing equal volumes of the feed solution entering the element and concentrate solution leaving the element.  $\pi_{\text{perm}}$  is the osmotic pressure of the permeate solution. Note that  $P$  approximates the average net driving pressure for an element. (To calculate the average net driving pressure,  $\pi_{\text{FeedAvg}}$   
30 would be replaced by the average osmotic pressure at the surface of the membrane.) This definition for standard specific flux does not include the impact of polarization on osmotic pressure, so that the term  $P$  slightly over-estimates the net driving pressure, but it is simpler to calculate.

This definition of an element's standard specific flux attempts to maintain as much consistency as possible with accepted industry practices while specifying a unique and easily determined parameter. The 24 hour time period for equilibration is common for specifying element performance. The test conditions (8% recovery, 25°C, and 32000 ppm NaCl) are consistent with tests used to characterize FilmTec's seawater products, and the other manufacturers in Table 2 specify similar recovery, temperature, and osmotic strength. The flows of commercial seawater elements in Tables 1 and 2 are all based on measurements made with 55.2 bar (800 psi) applied pressure, but defining an element's standard specific flux based on an 55.2 bar (800 psi) test would be problematic for the elements of this invention. Using a typical seawater test (55.2 bar applied pressure, 8% recovery, 25°C, and 32000 ppm NaCl feed) would result in exceeding maximum flux guidelines for elements with high standard specific flux. Also, very high fluxes cause excessive polarization and substantial internal resistances to flow, such as within the permeate carrier sheet (10), both effects causing the required pressure under more typical flux conditions (12-17 L/m<sup>2</sup>/hr) to be overestimated. For these reasons, an element's standard specific flux is defined in terms of a test that stipulates an average flux of 27 L/m<sup>2</sup>/hr during the measurement process. However, when the flux of an element does not exceed 34 L/m<sup>2</sup>/hr (20 gfd) in a typical seawater test, the standard specific flux can be reasonably estimated from the results (average flux and term *P*) of that test. For a well rejecting element with minimal pressure drop down the feed spacer (4), the term *P* above corresponds to approximately 27.6 bar (400 psi) in a typical 55.2 bar (800 psi) seawater test. A 32.5 m<sup>2</sup> (350 ft<sup>2</sup>) element with 21.2 m<sup>3</sup>/day (5600 gpd) would then have a flux of 27 L/m<sup>2</sup>/hr (16 gfd) and correspond to a standard specific flux of approximately 1.0 L/m<sup>2</sup>/hr/bar (0.04 gfd/psi).

A publication ("FILMTEC™ Membranes: How to evaluate the active membrane area of seawater reverse osmosis element", Form No. 609-00434-803, FilmTec Corp., Edina, MN, 12/01/03) provides a procedure for accurately measuring the active area of an element. If one assumes that there are 32.5m<sup>2</sup> (350 ft<sup>2</sup>) of membrane in elements of Table 2, estimates for their standard specific flux vary between about 0.75 L/m<sup>2</sup>/hr/bar (0.03 gfd/psi) and 1.25 L/m<sup>2</sup>/hr/bar (0.05 gfd/psi). Independent of the exacting details, the elements of Tables 1 and 2 fall within a narrow range of standard specific flux. Approximately 1.0 L/m<sup>2</sup>/hr/bar (0.04 gfd/psi) for seawater elements has been the norm since commercial introduction of aromatic polyamide membranes despite the observations that 1) membranes with high specific flux have been available since the introduction of aromatic

polyamide membranes, 2) elements having substantially higher standard specific flux are currently sold into other applications, and 3) low pressure RO/NF applications have been focused on increasing standard specific flux for the last 20 years.

A standard specific flux for an element type may be defined as the mean value of standard specific flux for a representative population of individual elements (at least 20) belonging to that type (or class or model). In this invention, it is required that at least two, and preferably three, individual elements within a vessel have substantially different values of standard specific flux, but it is even more desirable that at least two, and preferably three, elements within a vessel belong to different recognized classes (or types), such that the standard specific flux of these different element types vary substantially. As previously mentioned, elements of the same type are similarly constructed and are supplied by the manufacturer with a separate designation corresponding to the group of elements of that type. In the past, it has been known to alleviate the impact on system performance of the natural variability within an element type by testing elements, sorting elements according to flow, and providing a loading plan that specifies the position of specific individual elements (as by serial number) within each vessel. However, specifying the use of more than one type of element within a vessel permits a wider range of standard specific fluxes to be used. Specifying the type of element at each position within a vessel also allows for a substantially simplified loading process for large systems with several parallel vessels. At the same time, tight tolerances can be maintained for the standard specific flux of elements at any one position in a vessel. It is most preferable that individual elements used in this invention have a standard specific flux that deviates from the standard specific flux for the corresponding element type by less than 20%. Even more preferable is that this deviation is less than 15%.

As previously mentioned, a key reason that membranes having 1.0 L/m<sup>2</sup>/hr/bar (0.04 gfd/psi) have become the defacto standard for seawater applications is the imbalance in flux that results within a vessel of multiple elements. This imbalance is particularly acute when elements having a high standard specific flux are operated in feed solutions with high osmotic strength, such as seawater. At reasonable recoveries, high flux on a lead element (50) results in concentration of the feed solution, and the increased osmotic strength of the feed causes subsequent elements in the vessel to operate at substantially lower flux. This can be seen in the Comparative Examples III and IV to be presented for 50% recovery of seawater. A vessel of elements having standard specific flux of 0.98 L/m<sup>2</sup>/hr/bar (0.04 gfd/psi) showed extremes of operating fluxes ranging between 5 L/m<sup>2</sup>/hr and

34 L/m<sup>2</sup>/hr. However, in a similar simulation with elements of standard specific flux 1.19 L/m<sup>2</sup>/hr/bar (0.07 gfd/psi), an even lower flux of 3.3 L/m<sup>2</sup>/hr/bar (1.9 gfd) results near the outlet end (48) and a much too high flux of 44 L/m<sup>2</sup>/hr (26 gfd) results near the inlet end (46) of the vessel.

5           The problems of high flux are well recognized in the industry. The elements in Table 2 all have flows near 23 m<sup>3</sup>/day (6000 gpd) during a typical seawater test. This is consistent with the common range of 55-69 bar (800-1000 psi) for seawater operation. For typical osmotic strengths, a 23 m<sup>3</sup>/day seawater element with 32.5 m<sup>2</sup> (350 ft<sup>2</sup>) of active area can be guaranteed to operate at 55 bar (800 psi) with less than 34 L/m<sup>2</sup>/hr (20 gfd), an  
10 often quoted upper bound for flux. Fouling over time and differences in temperature and osmotic strength may potentially allow the same element to stay within these limits at even 68.9 bar (1000 psi) or for some conditions even 82.7 bar (1200 psi). The two highest flow elements in Tables 1 and 2 are both from FilmTec. By maintaining an element specification of less than 29 m<sup>3</sup>/day (7600 gpd), FilmTec is assured that a 35.3 m<sup>2</sup> (380 ft<sup>2</sup>) lead element  
15 will operate at less than 34 L/m<sup>2</sup>/hr (20 gfd) under typical seawater test conditions. FilmTec's 9000 gpd element (SW30-380) would commonly exceed FilmTec's own maximum flux guidelines if used to desalinate seawater in the typical fashion, but this element has been aimed at specialized applications, as described in its product information sheet (Form No. 609-00242-703, FilmTec Corp., Edina, MN, 12/18/03).

20           This invention will allow for acceptable use of standard size seawater elements with even higher flows. The invention necessitates that there be more than one type of element within a vessel. In particular, the standard specific flux for a downstream element in the vessel is at least 50% greater, and more preferably at least 100% greater, than the standard specific flux for an upstream element in the same vessel. It is most preferred that this  
25 downstream element have a standard specific flux greater than 1.5 L/m<sup>2</sup>/hr/bar (0.061 gfd/psi).

It is important to this invention that elements having high standard specific flux also maintain a relatively low solute permeability coefficient. A solute permeability coefficient, often referred to as a B-value, determines the rate at which salt diffuses through a  
30 membrane and has been discussed in references such as (Osada & Nakagawa, *Membrane Science and Technology*, Chapter 9, "Reverse Osmosis," Marcel Dekker, Inc., New York, 1992). The solute permeability coefficient is one of the fundamental parameters of an osmotic membrane but its value is known to vary with solute composition and particularly

with temperature. For a specific set of operating conditions, the solute permeability coefficient of a membrane  $B_{mem}$  may be calculated from flux  $J$  and the intrinsic solute passage  $C_p/C_m$ , according to the following formula:

$$B_{mem} = ( J * C_p / C_m ) / ( 1 - C_p / C_m )$$

- 5 In this equation,  $C_p$  and  $C_m$  represent the concentrations of solute in the permeate and at the membrane's surface on the feed side, respectively. The concentration at the membrane's surface may be greater than the concentration in the feed due to concentration polarization.

Within this specification, a standard solute permeability for an element is defined similarly, but with the additional requirement that parameters are obtained under the test  
10 conditions specified for the standard specific flux. Specifically, measurements are made after 24 hours of operation using 25°C, 32000 ppm NaCl in the feed, 8% recovery, and a flux of 27 L/m<sup>2</sup>/hr. Additionally, the standard solute permeability  $B_{ele}$  is defined in terms of an average feed concentration,  $C_f$ , corresponding to the concentration of NaCl obtained by mixing equal volumes of the feed solution entering the element and concentrate solution  
15 leaving the element. While the simplified formula does not account for the impact of concentration polarization, it does allow for more facile measurement of required parameters.

$$B_{ele} = ( J * C_p / C_f ) / ( 1 - C_p / C_f )$$

In this specification, the standard solute permeability is used to compare different elements  
20 by ratio. While the standard solute permeability of an element is defined based on specific test conditions, it is possible to approximate this ratio for two elements, provided that temperature, concentrations, and recovery are similar for the two tests and provided that measured flux in the tests are less than 34 L/m<sup>2</sup>/hr (20 gfd).

The last element in series within a vessel operates with higher salt concentrations in  
25 the feed than other elements, so poor rejection can strongly influence system performance and it is important that salt rejection of the tail element be high. It is preferable that the tail element has at least sufficiently good salt rejection to produce potable water (< 500 ppm) when operating on its own during a standard test with 25°C, 32000 ppm NaCl in the feed, 8% recovery, and a flux of 27 L/m<sup>2</sup>/hr. Element C in Table 3 had a permeate concentration  
30 of 357 ppm under conditions used to measure standard solute permeability, and the three element vessel including that element in Example I resulted in an even flux distribution and produced potable water. More preferably, the rejection should be of bottled water quality

(< 300 ppm) in this test. Element E in Table 3 had a standard specific flux similar to that for Element C, but its standard solute permeability corresponds to about 237 ppm in a test with 25°C, 32000 ppm NaCl in the feed, 8% recovery, and a flux of 27 L/m<sup>2</sup>/hr.

This element would allow potable water to be produced with even lower pressures or higher recoveries. In the case of NF applications, substantial passage of NaCl (greater than 20% passage of NaCl with the above test conditions) is desirable for all elements within the vessel, but it is also desired that elements have high rejection of another component.

For seawater NF applications using elements of different standard specific flux within a vessel, it is preferable that the passage of sulfate be less than 1% for any element in the vessel when elements are tested individually on a feed consisting of 32000 ppm NaCl and 2000 ppm MgSO<sub>4</sub>, using conditions of 25°C, 8% recovery, and 27 L/m<sup>2</sup>/hr flux.

Feed spacers (4) are described in several patents and applications including US Patent application 20030205520 which is incorporated by reference. The feed spacer (4) of an element provides a path for feed flow across the membrane surface. It also creates mixing at the membrane surface that decreases concentration polarization. The cost of this enhanced mass transfer is increased pressure drop down the length the element, and the sum of pressure drops for individual elements in series produces a pressure drop down the vessel. While this pressure drop will tend to increase flux imbalance, the inventors recognize that mixing at the membrane surface is particularly important in the case of high osmotic strength solutions. For downstream elements, and particularly the last element in series (54), where osmotic strength is greatest and pressure drop has the least impact on vessel performance, it would be advantageous to provide greater mixing. It is within the scope of this invention that the lead element (50) and tail element (54) in the vessel may use different feed spacer materials and it is most preferred that the standard pressure gradient for the feed spacer (4) of the last element be at least 50% greater than the standard pressure gradient for feed spacer (4) of the first element. The standard pressure gradient for a feed spacer (4) is defined within this specification to be the pressure gradient (pressure drop per unit of distance) in the direction of feed flow measured by passing 25°C water through the element while permeate flow is prevented. Within this test, the volumetric flow rate of water is specified to be proportional to the active membrane area within the element and to be inversely proportional to the length of the element. For a one meter long, 35.3 m<sup>2</sup> (380 ft<sup>2</sup>) element, the flow rate used to measure the standard pressure gradient is 190 m<sup>3</sup>/day. It is most preferable that the standard pressure gradient be at least 0.4 bar/m, corresponding to a 6 psi pressure drop across a meter long element.

High recoveries, as allowed by our invention, can substantially reduce the volume of feed solution flowing across the membrane's surface. Decreased feed velocity increases polarization, decreases flux, and promotes fouling. It can also result in operating outside an element manufacturer's specifications for maximum recovery. For these reasons, it is  
5 recognized that elements may be staged within a vessel (40) so that the velocity of feed flow across the membrane surface is maintained high. In a preferred embodiment, a downstream element in the vessel (40) may have a feed spacer cross sectional area that is at least 15% smaller than that for the first (or lead) element (50), and more preferably 30% smaller. In this case, when the element operates according to a <15% recovery guideline, the  
10 velocity of feed across the last element in series (54) is greater than the velocity within the previous one or two elements in series. The feed spacer cross sectional area is calculated by multiplying the thickness of the feed spacer by half the active membrane area and dividing by the length of the element. Additionally, when such an element has an outer diameter that is substantially smaller than the inner diameter of a vessel, it is required that feed flow is  
15 prevented from bypassing the element.

It has been contemplated (US 4046685), and it is within the scope of this invention, to remove permeate flow from both ends (46,48) of the vessel (40) and segregate permeate generated in elements at opposite ends. As illustrated in Fig 4, in this case the vessel (40) has permeate ports (62,64) on both its ends to provide means for fluid to pass between  
20 external piping and the permeate tubes (6) of end elements (50,54). A barrier (66) to permeate flow segregates the two permeate streams that leave from elements at opposite ends of the vessel. The barrier (66) is located between elements or within the permeate tube (6) of one element, and it prevents the two permeate streams from substantial mixing. This is shown in Fig. 4. It is not required that the barrier to flow be impenetrable to prevent  
25 substantial mixing, but the barrier should have a resistance to flow that exceeds by at least a factor of five the resistance to flow of permeate interconnectors (58) used within the vessel (40) to connect the permeate tubes of adjacent elements. In the conventional design, this segregation of permeate streams allows the best quality permeate to be removed from the upstream elements in a vessel (40). At the same time, using downstream elements of higher  
30 standard specific flux can allow for a high flow of permeate from the tail end (48) of the vessel (40) having good permeate quality. Depending on operating conditions, water from downstream elements may be suitable for industrial, potable, or bottled water. It is also possible to subject this permeate stream to additional treatment steps or to use it in blending.

When the barrier to permeate flow (66) is essentially impenetrable, the two permeate streams may also be maintained at different pressures. In this case, use of permeate back pressure can provide a relatively even flux distribution, independent of whether elements of higher standard specific flux are located near the upstream end (46) or downstream end (48) of the vessel (40). In a preferred embodiment, permeate back pressure results when the combined permeate stream from high standard specific flux elements becomes the feed stream for a second pass filtration vessel. Most preferably, elements of higher standard specific flux would be located near the inlet end (46) of the first vessel, as this arrangement provides a larger net driving pressure to cause permeate flow in both first and second pass elements. This arrangement can be used to provide a more uniform flux distribution within the first vessel.

A particular advantage of this approach is that this invention is conducive to upgrading existing systems. This invention provides a means to increase the recovery of an existing system with minimal capital expense. As existing large systems require greater productivity, higher recoveries may be attempted by increasing applied pressure, but this has limitations in fouling, energy requirements, and system design limits. Obtaining higher recoveries by replacing existing vessels and adding to the number of elements in series is generally not economical. Commonly, the only reasonable option has been to build additional parallel trains, with substantial cost in capital. This invention allows vessel efficiency to be increased by removing one or more existing elements from the vessel and loading new elements, at least one of which has a standard specific flux greater than  $1.5 \text{ L/m}^2/\text{hr}/\text{bar}$  ( $0.061 \text{ gfd/psi}$ ). Selectively replacing elements to provide a more even flux distribution can allow design guidelines to be maintained while operating a vessel at higher average flux and greater recovery. This is evidenced as a particularly advantageous option when one recognizes that elements typically represent only 5% of the capital cost in seawater installations. Alternatively, a vessel may be upgraded to obtain the same recovery while operating with a decreased maximum average element flux.

#### Examples

While not intending to limit the scope of invention, the present invention is further illustrated by the following examples of embodiments:

##### Example I

Four elements having  $2.6 \text{ m}^2$  of active membrane area were constructed using a membrane that will be commercialized in 2004 in a 20 cm diameter standard size, as

FILMTEC SW30XLE-380. Three of these elements were treated by contacting the membrane with 2000 ppm NaOCl for 30 minutes. The pH was 10.5 as described in US 5876602. Table 3 shows the measured standard specific flux and standard solute permeability for these four elements and for one 2.6 m<sup>2</sup> element made using membrane consistent with FILMTEC SW30HR-380 elements.

Table 3. Elements described in Example I.

Element	Membrane	Standard Specific Flux L/m <sup>2</sup> /hr/bar (gfd/psi)	Standard Solute Permeability L/m <sup>2</sup> /hr (gfd)
A	SW30HR	1.07 (0.043)	0.29 (0.17)
B	SW30XLE	1.43 (0.058)	0.45 (0.26)
C	SW30XLE (treated)	2.12 (0.086)	0.33 (0.19)
D	SW30XLE (treated)	1.85 (0.075)	0.32 (0.19)
E	SW30XLE (treated)	1.99 (0.082)	0.20 (0.12)

Elements A, B, and C were loaded into a vessel, so that element A was in the lead the position and element C was in the tail position. Permeate flow was blocked between elements B and C to allow the permeate solution from element C to be collected separately. With a 3.2% NaCl feed at 21°C, an applied pressure of 798 psi resulted in a combined recovery of 25.8%. The flux of elements A and B averaged 29 L/m<sup>2</sup>/hr (17 gfd). The flux of the tail element was 36.2 L/m<sup>2</sup>/hr (21.3 gfd). The concentration of NaCl in the combined permeate was 428 ppm.

#### Comparative Example I

Four elements having 2.6 m<sup>2</sup> of active membrane area were made with membrane consistent with FILMTEC SW30HR-380 elements. The elements had an average standard specific flux of 0.98 L/m<sup>2</sup>/hr/bar (0.04 gfd/psi) and an average standard solute permeability of 0.066 L/m<sup>2</sup>/hr (0.039 gfd). With a 3.2% NaCl feed at 21°C, an applied pressure of 55 bar (798 psi) resulted in a combined recovery of 19.6% from the four elements in series. The combined permeate concentration was 245 ppm. The flux of the lead element was not measured, but its standard specific flux suggests a value near 27 L/m<sup>2</sup>/hr. The average flux of first three elements was 22.6 L/m<sup>2</sup>/hr (13.3 gfd) and the average flux of the forth element in series was 15.6 L/m<sup>2</sup>/hr (9.2 gfd). Operating at less than 20% recovery, the flux of the forth element has been reduced to about 60% of the lead element flux. As compared to

Example I, a greater number of elements operating at the same applied pressure produced less water and had a less homogenous flux distribution.

#### Example II

FILMTEC SW30XLE-380 elements, similar to those that will commercially introduced in 2004, were contacted for 30 minutes with either 1500 ppm or 2000 ppm of NaOCl at pH 10.5 to result in elements having the standard specific flux and standard solute permeability values shown in rows H and I of Table 4. Rows F and G in the table correspond to untreated FILMTEC SW30HR-380 and SW30XLE-380 elements, respectively. The ratio of standard solute permeability to standard specific flux for the tail element (0.064) divided by the ratio of standard solute permeability to standard specific flux for the lead element (0.071) is less than 1. For all elements in Table 4, the standard pressure gradient was approximately 0.2 bar/m and the feed spacer cross sectional area was approximately 230 cm<sup>2</sup>.

Table 4. Elements described in Example II

Element	NaOCl	Standard Specific Flux L/m <sup>2</sup> /hr/bar (gfd/psi)	Standard Solute Permeability L/m <sup>2</sup> /hr (gfd)
F	0 ppm	0.96 (0.039)	0.068 (0.040)
G	0 ppm	1.43 (0.058)	0.042 (0.025)
H	1500 ppm	2.09 (0.085)	0.13 (0.076)
I	2000 ppm	2.66 (0.108)	0.08 (0.100)

The flows for elements F, G, H, and I were simulated in FilmTec's simulation program (ROSA, version 5.4) by adjusting the fouling factor of the SW30-380 element to 0.64, 1.05, 1.44, and 2.0. A vessel composed of three elements of type F, one element of type G, and three elements of type I was simulated by separately simulating each element and allowing the concentrate from each element to become the feed to the next element. A correction was made after each element to permeate concentrations because of the differing solute permeabilities for these elements, as compared to the SW30-380. The ratio of the standard solute permeability for the SW30-380 the simulation to the ROSA simulation of In this simulation, 3.5% seawater feed was composed of 19479 ppm Cl, 10460 ppm Na, 1450 ppm Mg, 2760 ppm SO<sub>4</sub>, 450 ppm Ca, and 400 ppm K.

Using 66.6 bar (967 psi) applied pressure and 196 m<sup>3</sup>/day (36 gpm) raw water feed flow at 25°C, simulation indicate a 60% recovery is achieved. Calculation showed the greatest flux to occur with the lead element, and this element had a flux of 33.6 L/m<sup>2</sup>/hr (19.8 gfd). The maximum recovery per element was 15%, consistent with the guidelines of several manufacturers. The permeate concentration was calculated as 295 ppm, well below that for potable water.

#### Comparative Example II

By the methods of example 3, a simulation has been performed in which 60% recovery is obtained with seven FILMTEC SW30HR-380 elements in series. In that case, the first element had an average flux of 39.6 L/m<sup>2</sup>/hr (23.3 gfd), a recovery of 17%, and the applied pressure was 74.2 bar (1076 psi).

#### Example III and Comparative Examples III and IV

While commercial programs such as ROSA may provide a more accurate calculation due to consideration of a variety of complicated factors (e.g. compaction due to pressure, resistance to flow in channels, interacting solutes), a simple simulation can also demonstrate the importance of this invention. In cases below, a vessel containing one meter long, 20 cm diameter elements is simulated by considering performance down the vessel in one inch increments. In each case, the applied pressure, feed concentration, and the flow into the first element are given, and performance (pressure drop, flux, and salt passage) within each increment is calculated. Results are propagated to successive one-inch long sections down the vessel.

Calculations assume a specific flux and solute permeability for the membrane within each element, and flux and salt passage for each increment are calculated according to standard formulas (Osada op.cit.). Polarization at the surface is estimated from feed velocity and flux according to equations provided for FilmTec elements in G. Schock & A. Miquel, "Mass transfer and pressure loss in spiral wound modules," *Desalination*, 65, (1987), 339-352) and values were chosen to equal the standard specific flux and standard solute permeability values noted above. Within this calculation, the osmotic pressure is determined from a linear interpolation between 26 bar for 3.5% seawater and 71 bar for 8.8% seawater. Pressure drop down in the feed channel is assumed to be linear with feed velocity, and equal to 0.24 bar (3.5 psi) for a one meter long, 35.3 m<sup>2</sup> (380 ft<sup>2</sup>) element with a 189 m<sup>3</sup>/day (50000 gpd) average feed flow.

Tables 5-7 below provide results of three simulations, each having 174 m<sup>3</sup>/day (46000 gpd) feed flow and 50% recovery of 3.8% seawater. Example III and Comparative Examples III and IV, all used a pressure vessel containing six 35.3 m<sup>2</sup> (380 ft<sup>2</sup>) elements, and the average flux for the vessel was 17 L/m<sup>2</sup>/hr (10 gfd). The membrane specific fluxes (A values) assumed for elements all correspond to ranges easily available. The membrane solute permeability values (B values) are assumed to be the same (0.068 L/m<sup>2</sup>/hr) for all elements within the vessel, although this value has minimal impact on flux over operating ranges being examined. Salt passage for permeate from each element will be roughly proportional to the assumed B value. The calculated combined permeate concentrations for Example III, Comparative Example III, and Comparative Example IV were 369 ppm, 315 ppm, and 369 ppm, respectively, all less than 500 ppm. Required pressures were 68.8 bar (998 psi), 72.5 bar (1051 psi), and 66.9 bar (971 psi), respectively. Example III requires low pressure, but also demonstrates that elements in a vessel may be run with low values for the maximum flux, average flux, and maximum element recovery. The flux distribution was more homogenous in Example III.

Table 5. Flux distribution for Example III

Element	Area (m <sup>2</sup> )	A value (Lmh/bar)	B value (Lmh)	Avg Flux (Lmh)	Max Flux (Lmh)	Recovery
1	35.3	0.739	0.068	23.1	24.8	11.5%
2	35.3	0.862	0.068	21.6	23.8	11.9%
3	35.3	1.231	0.068	21.1	24.4	13.1%
4	35.3	1.970	0.068	18.0	22.6	12.9%
5	35.3	2.462	0.068	11.7	15.1	9.7%
6	35.3	2.462	0.068	6.8	8.7	6.2%

Table 6. Flux distribution for Comparative Example III

Element	Area (m <sup>2</sup> )	A value (Lmh/bar)	B value (Lmh)	Avg Flux (Lmh)	Max Flux (Lmh)	Recovery
1	35.3	0.985	0.068	30.7	34.0	15.3%
2	35.3	0.985	0.068	24.1	27.2	13.8%
3	35.3	0.985	0.068	18.2	20.9	12.1%
4	35.3	0.985	0.068	13.2	15.4	10.0%
5	35.3	0.985	0.068	9.3	11.0	7.9%
6	35.3	0.985	0.068	6.5	7.6	5.9%

Table 7. Flux distribution for Comparative Example IV

Element	Area (m <sup>2</sup> )	A value (Lmh/bar)	B value (Lmh)	Avg Flux (Lmh)	Max Flux (Lmh)	Recovery
1	35.3	1.724	0.068	37.7	44.3	18.8%
2	35.3	1.724	0.068	25.6	30.9	15.4%
3	35.3	1.724	0.068	16.8	20.7	11.9%
4	35.3	1.724	0.068	10.7	13.2	8.6%
5	35.3	1.724	0.068	6.8	8.5	5.9%
6	35.3	1.724	0.068	4.2	5.3	3.9%

#### 5 Example IV

Calculations were performed as in Example III, but using a 167 m<sup>3</sup>/day (44000 gpd) feed of 3.5% seawater. An applied pressure of 79.3 bar (1150 psi) resulted in a simulated recovery of 60.8% for this vessel. In this case, seven elements within the vessel potentially differed in A values, B values and active area, as noted in the table. The combined permeate concentration is estimated at 448 ppm. Simulations show each element within this vessel to have low values for maximum flux, average flux and element recovery. Dividing the total permeate flow by the active membrane area provides an average flux for the vessel of 18.8 L/m<sup>2</sup>/hr (11.1 gfd). As with the other examples provided for single vessels, calculated results are also applicable to a filtration system having parallel vessels that used corresponding elements at each position.

Table 8. Flux distribution for Example IV

Element	Area (m <sup>2</sup> )	A value (Lmh/bar)	B value (Lmh)	Avg Flux (Lmh)	Max Flux (Lmh)	Recovery
1	35.3	0.616	0.068	26.5	28.2	13.8%
2	33.0	0.739	0.068	26.1	28.7	14.4%
3	33.0	0.862	0.068	23.3	26.3	14.9%
4	30.7	1.231	0.170	21.2	25.5	14.9%
5	30.7	2.462	0.170	17.3	22.7	14.3%
6	30.7	2.462	0.170	9.5	12.6	9.2%
7	30.7	2.462	0.170	5.3	7.0	5.6%

## WHAT IS CLAIMED IS:

1. An apparatus for purifying water comprising a first filtration vessel with opposing inlet and outlet ends and at least three spiral wound elements in series within said first filtration vessel; wherein said at least three elements in series define a lead element proximate to said inlet end of said first filtration vessel and a tail element proximate to said outlet end of said first filtration vessel; said at least three elements in series comprising an element having a maximum value of standard specific flux, an element having a minimum value of standard specific flux, and an element having an intermediate value of standard specific flux, wherein said maximum value of standard specific flux divided by said minimum value of standard specific flux is greater than 2 and said intermediate value of standard specific flux divided by said minimum value of standard specific flux is between 1.25 and 1.75.
2. The apparatus of claim 1 wherein said element having a maximum value of standard specific flux belongs to a first element type, said element having a minimum value of standard specific flux belongs to a second element type, and the standard specific flux for said first element type divided by the standard specific flux for said second element type is greater than 2.
3. The apparatus of claim 1 wherein the standard specific flux of said tail element divided by the standard specific flux of said lead element is greater than 2.
4. The apparatus of claim 3 wherein the standard specific flux for said tail element is greater than  $1.5 \text{ L/m}^2/\text{hr}/\text{bar}$ .
5. The apparatus of claim 3 wherein said lead element has a standard specific flux less than  $1.0 \text{ L/m}^2/\text{hr}/\text{bar}$ .
6. The apparatus of claim 3 wherein said tail element has a feed spacer cross sectional area that is at least 15% less than the feed spacer cross sectional area of said lead element.
7. The apparatus of claim 6 wherein said tail element has a feed spacer cross sectional area that is at least 30% less than the feed spacer cross sectional area of said lead element.
8. The apparatus of claim 3 wherein the feed spacer for said tail element has a standard pressure gradient greater than  $0.4 \text{ bar/m}$ .

9. The apparatus of claim 3 wherein the feed spacer for said tail element has a standard pressure gradient in the axial direction that is 50% greater than the standard pressure gradient in the axial direction for the feed spacer sheet of said lead element.

10. The apparatus of claim 3 wherein the ratio of standard solute permeability to standard specific flux for said tail element divided by the ratio of standard solute permeability to standard specific flux for said lead element is less than 2.

11. The apparatus of claim 3 where said tail element produces a permeate salt concentration of less than 500 ppm when tested using 25°C, 32000 ppm NaCl in the feed, 8% recovery, and a flux of 27 L/m<sup>2</sup>/hr.

12. The apparatus of claim 3 wherein said first filtration vessel is one of at least three parallel filtration vessels.

13. The apparatus of claim 1 further comprising a barrier to permeate flow within said first filtration vessel, said barrier defining first and second combined permeate streams, said first combined permeate stream comprising the entire of permeate from said lead element and said second combined permeate stream comprising the entire of permeate from said tail element, said barrier further preventing substantial mixing of permeate between said combined permeate streams.

14. The apparatus of claim 13 wherein said barrier is essentially impenetrable, said first combined permeate stream comprises the entire of permeate from said element having a maximum value of standard specific flux, said maximum value of standard specific flux divided by said minimum value of standard specific flux is greater than 2, and said first combined permeate stream becomes the feed stream to a second filtration vessel.

15. A process for purifying water comprising the steps of:

flowing a feed solution through a first pressure vessel containing at least three spiral wound elements in series, said at least three elements in series defining a lead element proximate to the feed inlet end of said vessel and a downstream element; wherein said at least three elements in series comprise an element having a maximum value of standard specific flux and an element having a minimum value of standard specific flux, and said maximum value of standard specific flux divided by said minimum value of standard specific flux is greater than 1.5;

applying pressure to said feed solution to cause permeate to pass through each element within said vessel, and

removing permeate and concentrate solutions from said vessel,

wherein the feed solution has an osmotic pressure greater than 20 bar at the inlet of said vessel.

16. The process of claim 15 wherein said element having a maximum value of  
5 standard specific flux belongs to a first element type, said element having a minimum value of standard specific flux belongs to a second element type, and the standard specific flux for said first element type divided by the standard specific flux for said second element type is greater than 2.

17. The process of claim 15 wherein the standard specific flux for said  
10 downstream element divided by the standard specific flux for said lead element is greater than 1.5.

18. The process of claim 17 wherein said downstream element has a standard specific flux that is greater than  $1.5 \text{ L/m}^2/\text{hr}/\text{bar}$ .

19. The process of claim 18 wherein the average net driving pressure for said  
15 lead element divided by the average net driving pressure for said downstream element is greater than 2.

20. The process of claim 17 wherein the difference in applied pressure and osmotic pressure at the inlet of said vessel divided by the difference in applied pressure and osmotic pressure at the outlet of said vessel is greater than 2.

21. The process of claim 20 wherein said downstream element is proximate to  
20 the outlet end of said vessel.

22. The process of claim 20 wherein said lead element is operated with an average flux that is less than twice the average flux for said vessel.

23. The process of claim 18 wherein said vessel contains at least five spiral  
25 wound elements in series, the volume of said concentrate solution produced is no more than twice the volume of said permeate solution produced, the average flux for said vessel is at least 70% of said average flux for said lead element, and said lead element has an average flux of between 10 and  $27 \text{ L/m}^2/\text{hr}$ .

24. The process of claim 23 where said average flux for said vessel is at least  
30 80% of said average flux for said lead element.

25. The process of claim 20 wherein said lead element is operated with an average flux less than  $34 \text{ L/m}^2/\text{hr}$  and said vessel is operated with an average flux greater than  $24 \text{ L/m}^2/\text{hr}$ .

26. The process of claim 25 wherein said concentrate solution has an osmotic pressure that is more than twice said osmotic pressure at the inlet.

27. The apparatus of claim 18 wherein said downstream element has a feed spacer cross sectional area that is at least 30% less than the feed spacer cross sectional area of said lead element.

28. The apparatus of claim 18 wherein said downstream element has a NaCl passage greater than 20% when said element is tested individually using a flux of  $27 \text{ L/m}^2/\text{hr}$ , 8% recovery, and a  $25^\circ\text{C}$  feed solution consisting of 32000 ppm NaCl in water, and wherein said downstream element has a sulfate passage less than 1% when tested individually using a flux of  $27 \text{ L/m}^2/\text{hr}$ , 8% recovery, and a  $25^\circ\text{C}$  feed solution consisting of 32000 ppm NaCl and 2000 ppm  $\text{MgSO}_4$  in water.

29. The process of claim 18 wherein said steps of claim 18 are preceded sequentially by the following actions:

operating said vessel by flowing a feed solution through said vessel, applying pressure, and removing permeate and concentrate solutions, said vessel during this operation containing an initial set of reverse osmosis elements, said initial set of reverse osmosis element consisting of elements having a standard specific flux less than  $1.25 \text{ L/m}^2/\text{hr}/\text{bar}$ , then

removing from said vessel at least one of said elements having a standard specific flux less than  $1.25 \text{ L/m}^2/\text{hr}/\text{bar}$ , and then

adding at least one subsequent element to said vessel, said subsequent element having a standard specific flux greater than  $1.5 \text{ L/m}^2/\text{hr}/\text{bar}$ .

30. The apparatus of claim 15 further comprising an essentially impenetrable barrier to permeate flow within said first filtration vessel, said barrier defining first and second combined permeate streams, said first combined permeate stream comprising the entire of permeate from said lead element and said second combined permeate stream comprising the entire of permeate from said tail element; wherein said first combined permeate stream becomes the feed stream to a second filtration vessel, and wherein said lead element divided by said minimum value of standard specific flux is greater than 1.5.

31. A process for purifying water comprising the steps of:

flowing a feed solution through a pressure vessel comprising at least three spiral wound reverse osmosis elements, said at least three spiral wound elements comprising an upstream element, a downstream element, and an intermediate element located between  
5 said upstream and said downstream elements, wherein the standard specific flux for said downstream element divided by the standard specific flux for said upstream element is greater than 2, and the standard specific flux for said intermediate element divided by the standard specific flux for said upstream element is between 1.25 and 1.75,

10 applying pressure to said feed solution to cause permeate to pass through each element within said vessel, and

removing permeate and concentrate solutions from said vessel,

wherein the difference in applied pressure and osmotic pressure at the inlet of said vessel divided by the difference in applied pressure and osmotic pressure at the outlet of said vessel is greater than 2.

15 32. The process of claim 31 wherein said downstream element has a standard specific flux greater than  $1.5 \text{ L/m}^2/\text{hr}/\text{bar}$ .

33. The process of claim 31 wherein said feed solution has an osmotic pressure greater than 20 bar at the inlet of said vessel.

Abstract of the Invention

The present invention pertains to an apparatus and method for treating a solution of high osmotic strength, especially seawater and solutions of greater than 20 bar osmotic pressure, by passing the solution through a vessel containing spiral wound reverse osmosis or nanofiltration elements. The vessel contains at least three elements in series and at least  
5 two of these elements have standard specific fluxes that differ by at least 50%.

The invention allows a more even flux distribution within a filtration system to be obtained, and it may advantageously be combined with variations in element construction and feed spacers.

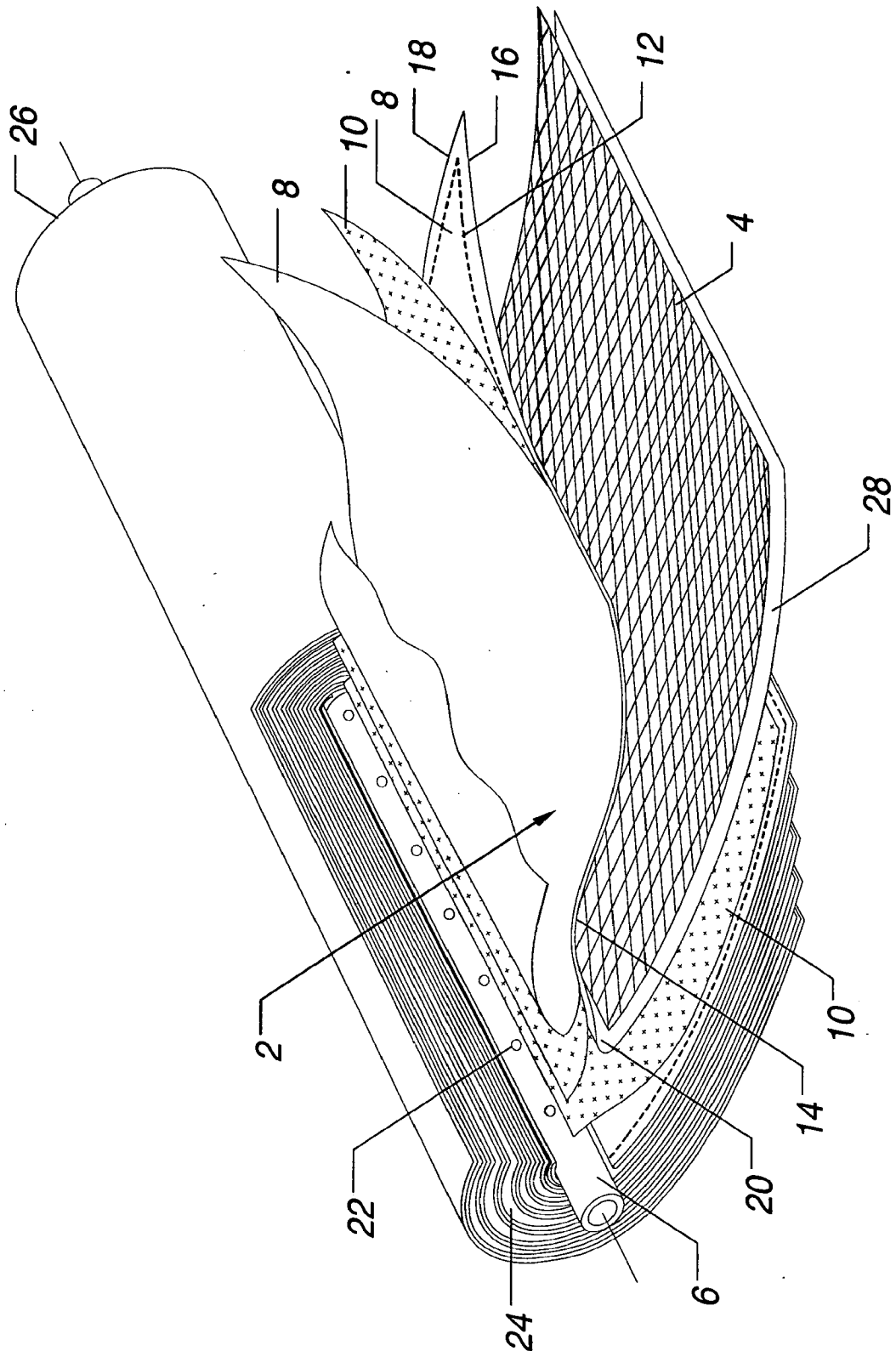
Title: APPARATUS FOR TREATING  
SOLUTIONS OF HIGH OSMOTIC  
STRENGTH

Inventors: William H. Mickols et al

Dow Case Number: 63549

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Fig. 1



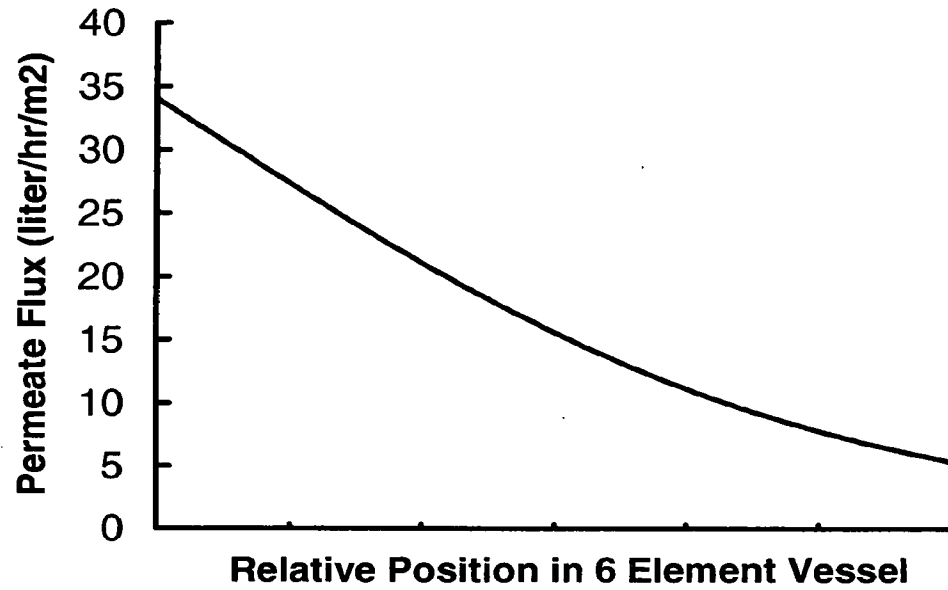
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" SOLUTIONS OF HIGH OSMOTIC  
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Fig. 2



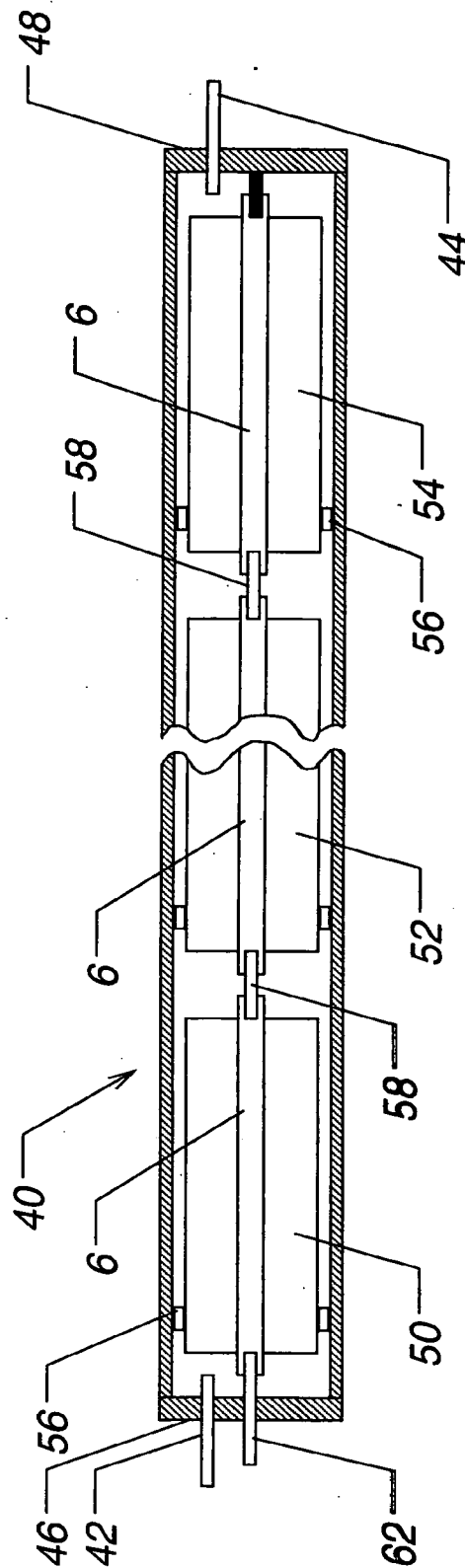
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Fig. 3



Title: APPARATUS FOR TREATING  
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Fig. 4

